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Performance of ceramic micro- and ultrafiltration membranes treating limed and partially clarified sugar cane juice

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ABSTRACT

The performance of ceramic membranes with pore sizes of 0.02, 0.05 and 0.10 μm in clarifying limed and partially clarified raw sugar cane juice was investigated under different operating conditions. For the 0.10 μm membrane, the increase in transmembrane pressure (TMP) from 1 to 3 bar increased the initial flux by 15.5% and increased the average flux over a period of 4 h by 11.9%. The initial flux of the 0.10 μm membrane increased dramatically at a TMP of 1 bar when the membrane underwent a chemical cleaning with 1% NaOH and NaOCl equivalent to 3000 ppm free chlorine for 1 h and the average flux over a period of 4 h was also increased. Among the three membranes tested 0.05 μm membrane performed better than the other two membranes and yielded higher initial and average fluxes. Out of the four fouling models used to fit the experimental data, the cake filtration model predicted the initial fluxes of 0.02 and 0.05 μm membrane more accurately. On the other hand, the combination of external and progressive internal fouling model predicted the performance of 0.10 μm membrane better compared to the others. Intermittent air back flushing improved the performance of 0.10 μm but did not have any effect on the performance of the other two membranes. However, all the membranes produced high quality filtered juice.

1. Introduction

The sugar industry needs to find efficient methods in clarifying the raw sugar cane juice in order to improve the quality of the clarified juice and to reduce or eliminate the usage of chemicals (lime). Conventional clarifiers use heavy equipments which lead to high operating costs and associated environmental problems. Further, the mud produced in a conventional clarifier is sent to a rotary vacuum filter to recover the sugar. However, the filtrate from the vacuum filter generally contains impurities and will not be able to enter the evaporation station directly and should be returned to the clarifier. This will increase the loading to the clarifier.

In sugar mills, ensuring the production of juice of consistently high clarity and low color through the clarification process is a challenging task. The variations in the incoming juice characteristics due to differences in cane variety, soil and growing conditions, weather patterns and season makes this task even more challenging. The membrane filtration promises superior quality juice with better clarity, much lower viscosity and noticeable color removal [1–3]. Ultrafiltration of clarified sugar cane juice can be done through spiral wound or flat sheet filtration system using polymeric membranes or tubular filtration system using ceramic membranes [4]. The filtrate from the membrane has an increase of 1.5–3 unit of juice purity, which is a remarkable improvement compared to the increase of 0.5–1 unit obtained in the liming-sulphitation process [5]. Membrane clarification yielded multi-fold improvement in juice clarity with nearly 60% reduction in color [6] as well as reduction in the inorganic contents of the juice.

In a study conducted with 0.02 μm ceramic membrane treating brown sugar solution of 28 °Brix at 60 °C produced 148 and 198 L/(m^2 h) of steady state fluxes at a TMP of 3 bar and cross flow velocities of 5.4 and 7.7 m/s, respectively. When the TMP was increased to 5 bar, the corresponding fluxes were 183 and 217 L/(m^2 h), respectively [7]. The steady state flux was reached within 10 min of the commencement of the experiments. Similarly, in another study, 230, 260 and 150 L/(m^2 h) of fluxes were obtained when treating 20 °Brix solution at 90 °C with 0.14, 0.20 and 0.45 μm membranes [8]. When 50 °Brix solution at 85 °C was treated with 0.1 μm membrane, the flux was around 50 L/(m^2 h) [9]. The 0.1, 0.2, 0.5, 0.8 and 1.4 μm membranes produced 38.0, 27.0, 30.0, 52.0 and 62.7 L/(m^2 h) of fluxes when treating 60 °Brix solution at 80–90 °C [10].

However, application of membranes in clarifying raw sugar cane juice to produce raw sugar using membranes is lacking in sugar mills. Thus, this study is aimed at investigating the performance of a laboratory scale ceramic membrane system in treating limed

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Table 1
Material characteristics and module details of the membrane system used in this study.

Item	Description
Manufacturer	Jiangsu Jiuwu HiTech, Nanjing, China
Membrane type	Tubular
Membrane material	ZrO_2
Membrane support material	α-Alumina oxide
Pore size	0.02, 0.05 and 0.1 μm
Pure water permeability	316 L/(m^2 h bar) (0.02 μm) 597 L/(m^2 h bar) (0.05 μm) 533 L/(m^2 h bar) (0.10 μm)
Porosity	35%
Length	500 mm
Number of channels	19
Channel diameter	4 mm
Surface area	0.1193 m^2

sugar cane juice. The aims of this study are to (i) evaluate the flux obtained for membranes with different pore sizes under different operating conditions such as transmembrane pressure (TMP) and air back flushing, (ii) evaluate the rate of fouling of membrane under the operating conditions mentioned above, (iii) selecting an appropriate mathematical model to predict the performance of the membranes and (iv) identifying appropriate chemicals for cleaning the fouled membrane and quantify the corresponding membrane resistance.

2. Materials and methods

Ceramic membranes with three different pore sizes (0.02, 0.05 and 0.10 μm) were used to filter the limed and partially clarified sugar cane juice under different operating conditions. The specifications of membranes are given in Table 1 and the operating conditions of each experimental run are given in Table 2. The experimental setup of the system is shown in Fig. 1.

Sugar cane variety Q200 was collected from Paluma (Queensland, Australia) and stored inside a large cold room at the Mechanical Engineering workshop of James Cook University at 10 °C for experiments. Raw sugar cane was crushed using the sugar cane miller. The raw sugar cane juice was then filtered through a 250-μm sieve to remove large fibers. Around 40 L of raw sugar cane juice was treated with $Ca(OH)_2$ and mixed by a stirrer to raise the pH from 5.2–5.5 to 7.5. The treated juice was kept unstirred for 1 h for the flocculated solid particles to settle. Supernatant juice was siphoned, filtered through a 125-μm sieve and diluted with DI water to adjust the sucrose content to be around 16 °Brix. The diluted solution (around 50 L) was then used as the feed for experiment. The above pre-treatment was used in order to simulate the conditions applied in sugar mills.

The juice volume in the feed tank was kept constant at 20 L throughout experiment by pumping the juice (30 L) from the limed juice tank with a peristaltic pump continuously. Sugar cane juice in the feed tank was maintained at 60 °C by a water bath. The juice was circulated through the membrane module by the centrifugal pump for juice filtration. The valves before (V1) and after (V2) the membrane module were adjusted to obtain the desired operating transmembrane pressure and crossflow velocity. The cross flow velocity was maintained at 3 m/s in all experiments. The retentate was recycled to the feed tank while the permeate was collected in a vessel placed on an electronic balance (Ohaus-CD33) connected to a computer that received weight data at 5 min intervals. To compute the flux, the weight was converted to volume based on specific weight of the permeate. For experiments with air back flushing mode in runs 5, 7 and 9, compressed air at 5 bar was applied from the permeate side to feed side to evaluate the effect of compressed air on membrane fouling. In this mode the filtration was conducted in a cycle of three periods controlled by solenoid valves and timers: (1) filtration period: 5 min during which the permeate valve (V3) was opened, compressed air valve (V4) and air exhaust valve (V5) were closed; (2) air back flushing period: 4 s with permeate valve and air exhaust valve closed and compressed air valve open; (3) air exhaust period: 4 s where the permeate valve and compressed air valves were closed and the air exhausted valve was opened. Each experimental run was conducted for 4 h. The flux generally reached a steady state condition in 4 h in all experiments and thus the results obtained in those experiments are comparable.

Samples from both the feed tank and permeate stream were collected at initial, after 30 min, 1, 2 and 3 h for analyses. The sample volume collected was recorded and added to the total filtrate volume calculated from the weight data for the flux computation.

After every experiment, the sugar cane juice is drained from the membrane system. The membrane was then rinsed with de-ionized water and then cleaned with chemicals as given in Table 2. Membrane resistance was checked before and after every experiment by measuring the pure water flux at 30 °C and at different TMPs.

3. Analytical methods

Brix is a measure of refractometric dry substance (RDS). The juice was filtered through Whatman filter paper and measured Brix by a digital refractometer (Palette PR-101, Atago).

The Pol is a measurement of the total polarized substances in the juice, which is used to represent the sucrose content in the juice. The dry lead method [11] was used to measure the Pol. A 2-g of subacetate of lead was mixed with 200 mL of juice and mixed thoroughly. The solution was let several minutes for the precipitate to settle and the supernatant was filtered through Whatman filter paper. A polarimeter (SQF-WXG4, Vanco) calibrated in sugar degree (°Z) was used to measure the Pol reading of the filtered supernatant. The

Table 2
Operating and cleaning conditions of the experimental runs.

Run no.	Membrane pore size (μm)	TMP (bar)	Air back flushing	Cleaning chemicals[a]	Cleaning time
1	0.10	1.0	No	2% HNO_3 followed by 2% NaOH	40 min each
2	0.10	2.0	No	2% HNO_3 followed by 2% NaOH	40 min each
3	0.10	3.0	No	2% NaOH, 1% NaOH + 3000 ppm free chlorine	1 h, 1 h
4	0.10	1.0	No	1% NaOH + 3000 ppm free chlorine	1 h
5	0.10	1.0	Yes	1% NaOH + 3000 ppm free chlorine	1 h[b]
6	0.05	1.0	No	1% NaOH + 3000 ppm free chlorine	1 h
7	0.05	1.0	Yes	1% NaOH + 3000 ppm free chlorine	1 h
8	0.02	1.0	No	1% NaOH + 3000 ppm free chlorine	1 h
9	0.02	1.0	Yes	1% NaOH + 3000 ppm free chlorine	30 min

[a] Cleaning was carried after each run; the membrane was first cleaned with pure water and then with the chemicals.
[b] Used cleaning solution was replaced four times by fresh cleaning solution during this cleaning.

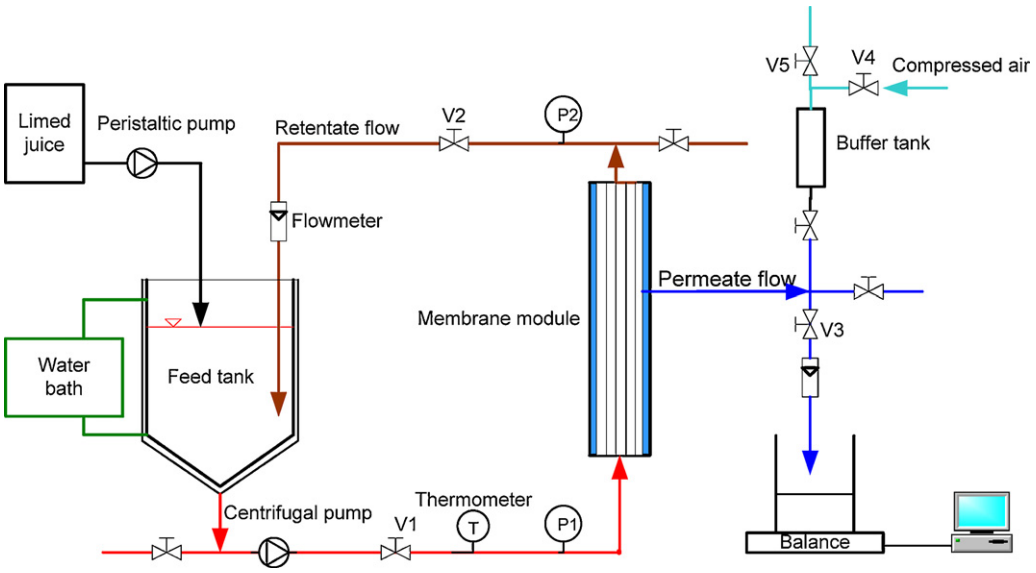

Fig. 1. Experimental setup to clarify limed sugar cane juice.

200 mm length pol tube was filled with the filtered supernatant to determine the Pol reading at 20 °C. Pol percent juice was calculated using the following equation:

$$\text{Pol percent juice} = \frac{\text{Pol reading}}{\text{Pol factor}} \quad (1)$$

where

$$\text{Pol factor} = \frac{100 \times \text{apparent density at } 20\,^{\circ}\text{C}}{26000} \quad (2)$$

$$\text{Purity of juice } (\%) = \frac{\text{Pol percent juice}}{\text{Brix}} \times 100 \quad (3)$$

$$\text{Purity rise} = (\text{Purity})_{\text{permeate}} - (\text{Purity})_{\text{feed}} \quad (4)$$

The color of the juice was measured according to GS1-7 method [12] where the sample was adjusted to pH 7 by 0.1N HCl and 0.1N NaOH and filtered through a 0.45 μm filter in order to measure absorbance at 420 nm using a spectrophotometer (HP-8453).

$$\text{Color (IU)} = \frac{10^8 \times A_s}{b \times \text{RDS} \times \rho} \quad (5)$$

where A_s is the absorbance at 420 nm, b is the cell length (cm), RDS is the refractometric dry substances (°Brix) and ρ is the density of the solution (kg/m^3).

The turbidity was measured according to GS 7-21 method [12]. Sample was measured at 900 nm using a spectrophotometer (HP-8453).

$$\text{Turbidity} = 100\ \frac{A_s}{b} \quad (6)$$

where A_s is the absorbance at 900 nm and b is the cell length (cm).

The pH of sugar cane juice solutions was measured by a digital pH meter (AQUA – pH, TPS).

4. Membrane cleaning

After each experiment the membrane was cleaned in place. It was first rinsed with de-ionized water for four to five times to remove all the juice from the system and until the resulting rinse produces clear water. Then chemical cleaning (for runs 1–8) was conducted at a TMP of 2 bar, cross flow velocity of 6 m/s and at a temperature of 40 °C. High cross flow velocity was employed in order to make sure best scouring of membrane surface occurs. During chemical cleaning, the permeate valve was closed for the first half duration of cleaning to remove the foulants from the membrane surface. For the rest of the cleaning duration, the permeate valve was opened to let the permeate to return to the feed tank. This helps in cleaning the foulants that are trapped under the membrane layer and support. Meanwhile, the cleaning in run 9 was modified to see the effect of membrane cleaning under higher temperature and lower TMP and cross flow velocity. Thus, the operating parameters during this cleaning were: TMP of 0.2 bar, cross flow velocity of 2 m/s and at a temperature of and 50 °C. Also, the permeate valve was opened during the entire duration of cleaning. Different chemicals and duration were applied in chemical cleaning of membranes. After cleaning with chemical, the membrane was rinsed with de-ionized water twice. When the cleaning was completed, fluxes at 30 °C and at different TMPs were measured to determine the membrane resistance.

5. Results and discussion

5.1. Flux

All the experiments were conducted for 4 h and Table 3 shows the initial, final and average fluxes obtained in all 9 runs along with the average fluxes during 1st, 2nd, 3rd and 4th hour of each run. The average flux was obtained using the permeate volume collected in 4 h and other flux values at a given period of time was computed using the volume of permeate collected during that time. The initial flux was computed using the volume of permeate obtained in the first 5 min since significant amount of permeate could be obtained in that first 5 min, which avoided any errors in the measurement of permeate volume and produced stable values for the initial flux. The initial fluxes obtained during the first three runs using 0.1 μm membrane were below 100 L/(m^2 h). Increasing the TMP to 2 bar in run 2 increased the initial flux from 55.6 (obtained in run 1) to 87.9 L/(m^2 h). But when the TMP was increased to 3 bar in run 3, the initial flux did not increase further but stayed at around 64.2 L/(m^2 h). Although the same cleaning procedure was carried out after runs 1 and 2, cleaning was not effective after run 2 (in which one would expect more fouling due to higher flux) and the membrane recovery was lower which resulted in lower initial flux in run 3. Therefore, the normalized flux at 1 and 3 bar of TMP were almost identical but the values at 2 bar are noticeably lower. Thus, a different cleaning procedure was introduced after run 3 to increase

Table 3
Fluxes observed in experimental runs.

Run no.	Initial flux (L/(m^2 h))	Final flux (L/(m^2 h))	Average flux (L/(m^2 h))	Average flux (L/(m^2 h))			
				1st hour	2nd hour	3rd hour	4th hour
1	55.6	30.9	35.2	44.5	33.9	31.6	30.7
2	87.9	31.4	41.4	56.4	40.6	36.4	32.1
3	64.2	30.9	39.4	49.0	39.3	37.0	32.2
4	140.7	26.6	46.0	78.4	42.7	34.2	28.9
5	146.4	36.6	56.6	89.8	55.6	43.0	38.0
6	165.4	44.7	65.6	100.6	63.1	51.6	47.1
7	149.6	39.0	59.2	87.6	58.1	49.0	42.0
8	135.0	36.6	59.4	92.2	60.1	47.2	38.0
9	124.7	37.6	53.7	79.0	54.3	42.3	39.2

the initial flux. Fig. 2(a) shows the decline in flux in those three runs. Fig. 2(b) shows the normalized fluxes for those runs in order to compare the performance of 0.1 μm membrane under different TMPs. It can be seen from Fig. 2(b) that the normalized flux obtained at TMPs of 1 and 3 bar is similar but an increase of 11.9% in the average flux can be achieved if the TMP is increased from 1 to 3 bar.

Runs 4 and 5 were conducted to see the performance of 0.1 μm membrane with and without air back flushing. In those runs, the initial flux, was more than 140 L/(m^2 h). This can be attributed to the change employed in chemical cleaning. At the end of runs 1 and 2, the membrane was cleaned with 2% HNO_3 followed by 2% NaOH as detailed in Table 2. But, at the end of run 3, the membrane was cleaned with 2% NaOH; before and after the run 4, it was cleaned with 1% NaOH and 3000 ppm chlorine. Fig. 3 shows the normalized flux obtained during runs 4 and 5 and it can be seen from Table 3 that an increase of 23% in the average flux can be achieved in air back flushing is applied to membrane once in every 5 min for 4 s. The flux values obtained in this study is comparable to the results obtained by Jacob and Jaffrin [13]. They used ceramic membranes with 15 kDa (ultrafiltration) and 0.10 μm pore sizes (microfiltration) and filtered 30 °Brix brown sugar solution at 60 °C with a cross flow velocity of 5 m/s and a TMP of 1 bar. The 0.1-μm ceramic membrane produced much larger initial flux (140 L/(m^2 h)) compared to the 15 kDa ceramic membrane (60 L/(m^2 h)). There was also less flux decline in microfiltration than in ultrafiltration. In the microfiltration the flux decreased from 140 to 100 L/(m^2 h) compared from 60 to 22 L/(m^2 h) in the ultrafiltration, in 8 h.

Runs 6 and 7 were conducted to see the performance of 0.05 μm membrane with and without air back flushing (Fig. 4) and runs 8 and 9 were conducted to see the performance of 0.02 μm membranes with and without air back flushing (Fig. 5). Unlike for 0.1 μm membrane, air back flushing did not improve the average flux, rather it decreased the average flux slightly. Thus, air back flushing is suitable for 0.1 μm membrane in order to increase the flux, while it did not affect the performance of 0.05 and 0.02 μm membrane.

From the above flux studies, it can be concluded that the ceramic membrane with a pore size of 0.05 μm can be employed to clarify

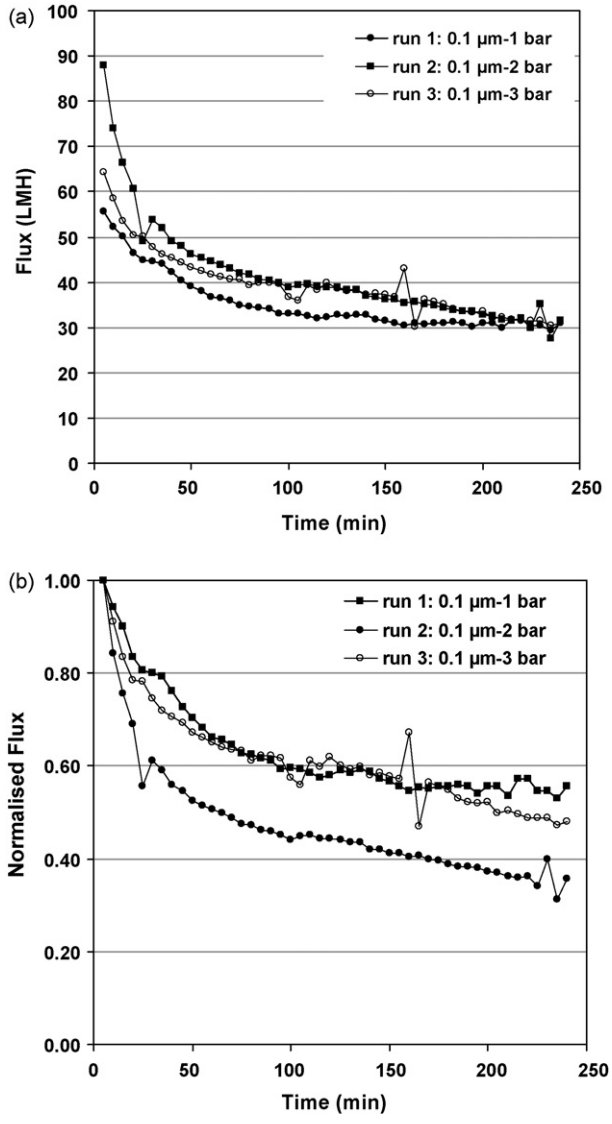

Fig. 2. (a) Temporal variation of flux for 0.1 μm membrane at TMPs 1, 2 and 3 bar. (b) Temporal variation of normalized flux for 0.1 μm membrane at TMPs 1, 2 and 3 bar.

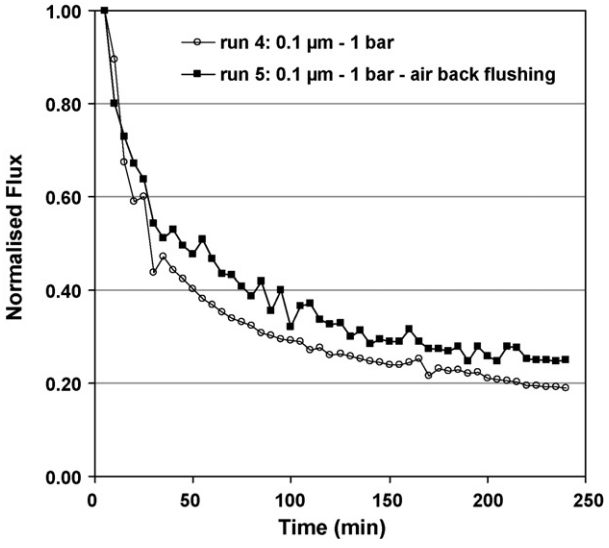

Fig. 3. Temporal variation of normalized flux for 0.1 μm membrane with and without air back flushing at a TMP of 1 bar.

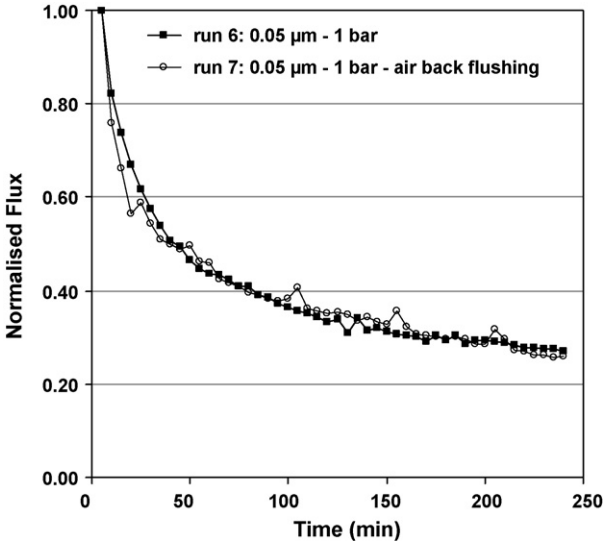

Fig. 4. Temporal variation of normalized flux for 0.05 μm membrane with and without air back flushing at a TMP of 1 bar.

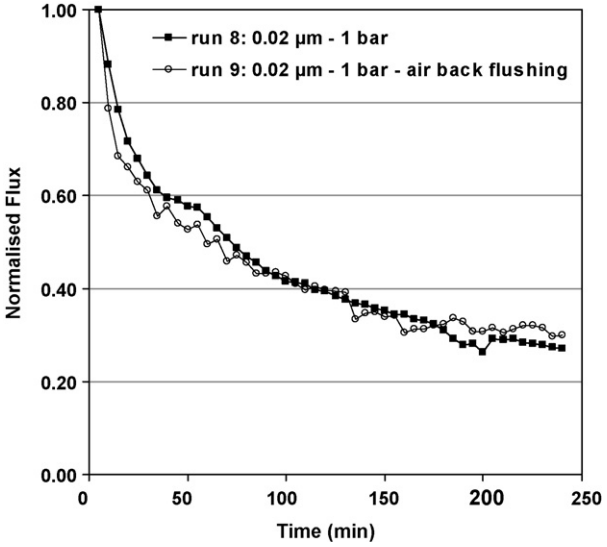

Fig. 5. Temporal variation of normalized flux for 0.02 μm membrane with and without air back flushing at a TMP of 1 bar.

sugar cane juice at an average flux of 65.6 L/(m^2 h), over a period of 4 h at a TMP of 1 bar. This flux is 16% more than the flux obtained from 0.1 μm membrane with back flushing and 10% more than the flux obtained from 0.02 μm without back flushing. This will become significant in large-scale operations and would reduce the membrane area required for a specified output of clarified sugar cane juice if 0.05 μm membranes are selected over 0.02 or 0.10 μm membranes. However, further studies are warranted to confirm this outcome.

Table 5
The performance of membranes in increasing purity and reducing turbidity and color of sugar cane juice.

Run no.	Purity rise	Reduction in turbidity (%)	Reduction in color (%)
1	1.7	99.6	42.6
2	1.8	99.7	49.9
3	2.2	99.8	55.2
4	1.7	99.7	38.9
5	2.1	99.6	41.2
6	2.3	99.7	44.8
7	2.4	99.7	40.4
8	2.3	99.7	44.0
9	2.1	99.6	42.1

5.2. Clarified sugar cane juice quality

The typical quality parameters of feed and permeate (filtered) streams of run 6 are given in Table 4. It can be seen from the table that the average values of turbidity and color of the feed stream generally tend to increase with time due to concentration but the Brix and pol of the feed stream did not change much. However, the permeate stream had consistent quality. Thus, all the runs showed more than 1.7 unit of purity rise, 99.6% reduction in turbidity and 38.9% reduction in color as shown in Table 5.

From the first three runs, it can be seen that higher the TMP the better the increase in purity, percentage reduction of turbidity and percentage reduction of color. Thus, higher TMPs could improve the quality of the clarified sugar cane juice. Again, under similar operating conditions, membranes with 0.02 and 0.05 μm pore sizes produced better quality clarified juice compared to 0.10 μm membrane. However the quality of the clarified juice obtained from all three membranes are comparable.

5.3. Membrane resistance and fouling models

The membrane resistance can be calculated using the following equation:

$$J = \frac{\Delta P}{\mu R_t} \tag{7}$$

where J is the permeate or clarified juice flux (L/(m^2 h)), ΔP is the transmembrane pressure (Pa), μ is the dynamic viscosity of the permeate (Pa s) and R_t is total membrane resistance (m^{-1}). The R_t has following components: the intrinsic membrane resistance, R_{m0}; the cake layer resistance, R_c; the fouling resistance, R_f. Generally, the entire cake layer resistance as well as most of the fouling resistance can be removed at the end of the filtration cycle by chemical cleaning.

The intrinsic membrane resistance was measured before the commencement of run 1 and at the end of all the runs as shown in Table 6. The 0.10 μm membrane was chemically cleaned before runs 1, 2 and 3 with 2% HNO_3 solution for 40 min followed by 2% NaOH for another 40 min. Although the intrinsic resistance of the membrane, R_{m0} returned closer to the initial R_{m0}, they were in the range of 3.87×10^{12} to 4.12×10^{12} m^{-1}, which seems to be 10-fold higher compared to the standard intrinsic resistance value

Table 4
Typical quality parameters of feed and permeate (filtered) streams obtained for run 6.

Time (h)	Brix (%)		Pol (%)		Purity (%)		Turbidity		Color (IU)		pH	
	Feed	Filtered	Feed	Filtered	Feed	Filtered	Feed	Filtered	Feed	Filtered	Feed	Filtered
0	16.6	16.3	14.3	14.3	86.1	87.7	205.7	0.4	14,290	9880	7.41	7.32
0.5	16.7	16.2	14.1	14.1	84.4	87.0	256.2	0.7	15,900	9180	7.42	7.26
1.0	16.6	16.0	14.1	13.9	84.9	86.9	274.4	0.8	17,770	8500	7.39	7.31
2.0	16.6	15.9	14.2	14.1	85.5	88.7	303.2	1.3	17,250	8710	7.38	7.35
3.0	16.9	16.1	14.2	13.9	84.0	86.3	298.3	0.8	16,930	8590	7.35	7.25

Table 6
Membrane resistance, before and after the clarification of sugar cane juice.

Run	Before run ($\times 10^{12}$ m^{-1})	After run ($\times 10^{12}$ m^{-1})		Recovery (%)
		After rinsing with pure water	After chemical cleaning	
1	3.87	7.22	3.19	121.2
2	3.19	12.40	4.12	77.4
3	4.12	14.70	5.56	74.1
4	0.78	6.51	0.80	96.7
5	0.80	6.35	0.87	92.1
6	0.60	5.09	0.74	81.2
7	0.74	4.62	0.79	93.5
8	1.16	5.18	1.29	90.4
9	1.29	5.20	1.40	92.0

of a ceramic membrane. The intrinsic resistance of the membrane was 5.56×10^{12} m^{-1} when it was cleaned with 2% NaOH alone for 1 h at the end of run 3. Thus, 1% NaOH and 3000 ppm free chlorine was used to clean the membrane for 1 h before commencing run 4. This brought the intrinsic resistance of the membrane down significantly (to 0.78×10^{12} m^{-1}).

The initial flux of the 0.1 μm membrane increased dramatically to 140.7 L/(m^2 h) at a TMP of 1 bar when the membrane underwent a chemical cleaning with 1% NaOH with 3000 ppm free chlorine for 1 h as mentioned above and the average flux over a period of 4 h was also increased to 46 L/(m^2 h). From the initial and average flux values obtained for each runs, it is evident, that cleaning the membrane with 1% NaOH and 3000 ppm chlorine solution for 1 h could remove most of the foulants from the membrane. The percentage recovery of membrane shown in Table 6 was calculated as the percentage ratio of the membrane resistance before the run and after the chemical cleaning.

There are several models that could describe the performance of membrane and some of them are [13,14]:

- Cake filtration model.
- Pore narrowing model (progressive internal fouling).
- Combination of external and progressive internal fouling.
- Complete pore blocking model.

The literature suggests that the first two models describe filtration of brown cane sugar solution well [13]. Some details of the above models are discussed below in order to understand the mechanisms involved in fouling of membranes.

5.3.1. Cake filtration model

When sugar juice is filtered through a membrane, a cake layer would form on the surface of the membrane due to the rejection of macrosolutes by the membrane. Thus, the resistance to filtration by this cake layer is assumed to increase proportionally with the volume of sugar juice filtered. Thus, the total resistance to filtration, R_t could be expressed as:

$$R_t = R_{m0} + \frac{\alpha C_w V_f}{A_o} \tag{8}$$

where R_{m0} is the intrinsic membrane resistance, α is the specific cake resistance per unit mass, C_w the concentration of rejected particles, V_f is the volume of filtered juice and A_o is the total membrane surface area. A relationship between the filtration time, t and the volume of juice filtered (V_f) could be obtained as follows:

$$\frac{t}{V_f} = \frac{1}{Q_o} + \frac{\alpha C_w V_f}{2A_o R_{m0} Q_o} \tag{9}$$

where Q_o is the initial flow rate of clarified juice (or permeate). Thus, experiments could be conducted under different experimental conditions to estimate α and C_w which could then be used to design large-scale membrane systems.

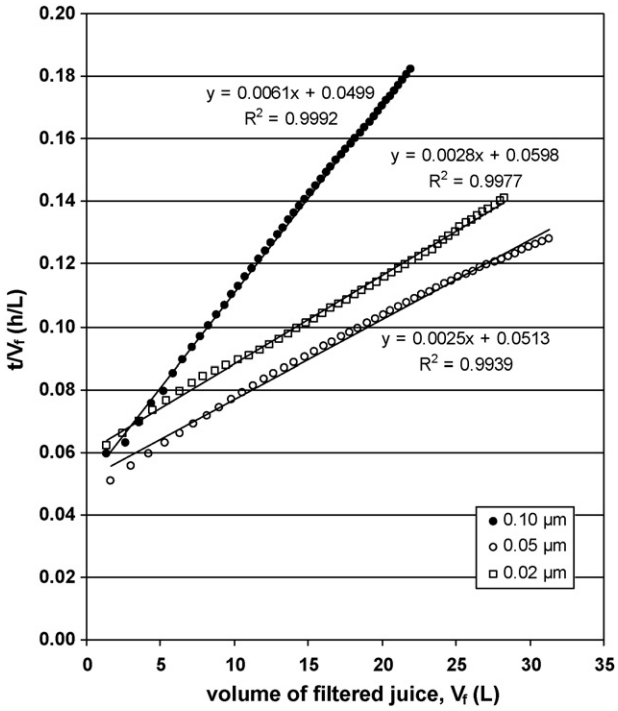

Fig. 6. Cake filtration model fitting to the experimental data obtained in runs 4, 6 and 8 (TMP = 1 bar; cross flow velocity = 3 m/s).

Fig. 6 shows the cake filtration model fitting to the experimental data obtained in runs 4, 6 and 8. While this model fits well with the data obtained in all three runs mentioned above, the initial flow rate or flux predicted by the model fits well when the pore size of the membrane becomes smaller. This indicates that the cake filtration model is much suitable for ultrafiltration membranes (0.02 and 0.05 μm) but could also be used to predict the performance of microfiltration membrane (0.10 μm) with reasonable accuracy as shown in Table 7.

5.3.2. Pore narrowing model

This model assumes that the membrane pores are progressively narrowed due to the penetration of macrosolutes inside the pores. Thus, the rate of reduction of pore radius, r could be expressed as:

$$2\pi NLr\left(\frac{dr}{dt}\right) = -CQ_f \tag{10}$$

where N is the number of pores on the membrane surface, L is the thickness of the membrane, C is the dimensionless parameter characterising the fraction of solute which gets adsorbed, Q_f is permeate flow rate at time, t. This would yield an expression:

$$\frac{1}{Q_f^{0.5}} = \frac{1}{Q_o^{0.5}} + \frac{CQ_o^{0.5}\, t}{V_p} \tag{11}$$

where V_p is the initial pore volume $= \pi N r_o^2 L$ where r_o is the initial pore radius. Again, experiments would determine C and would help to simulate the performance of the membrane.

Table 7
Cake filtration model fitting to the experimental data.

Run no.	Membrane pore size (μm)	Initial flux (L/(m^2 h))			αC_w (m^{-2})
		Experimental	Model	Error (%)	
4	0.10	140.7	168.0	19.4	2.263×10^{10}
6	0.05	165.4	163.4	−1.2	6.961×10^{9}
8	0.02	135.0	140.2	3.8	1.298×10^{10}

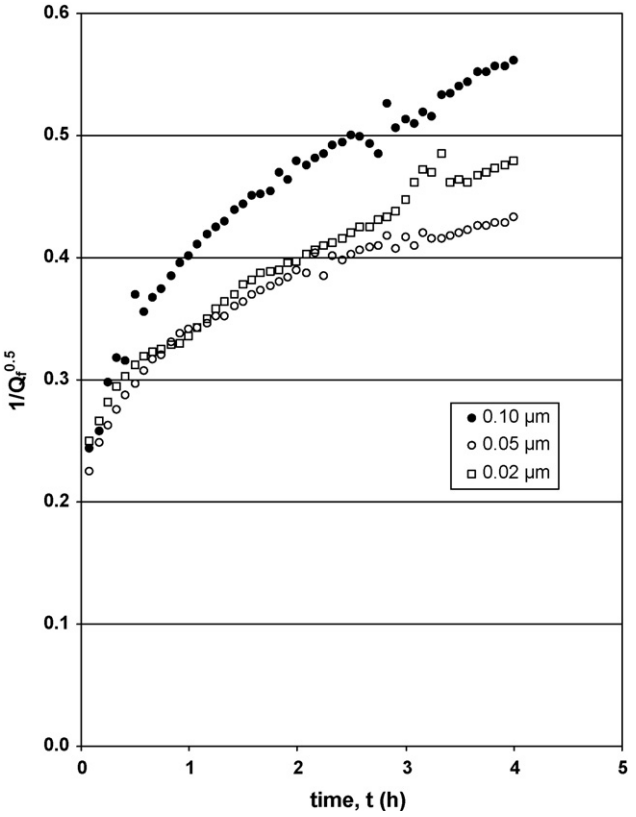

Fig. 7. Pore narrowing model fitting to the experimental data obtained in runs 4, 6 and 8 (TMP = 1 bar; cross flow velocity = 3 m/s).

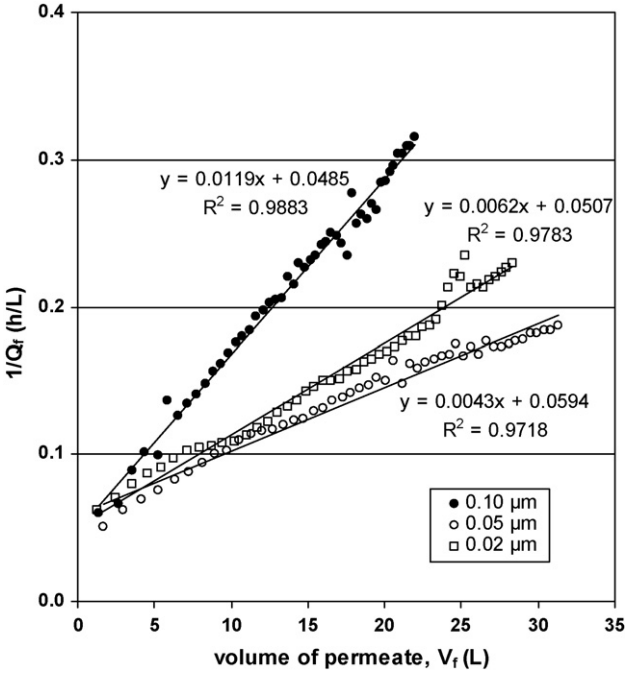

Fig. 8. Combination of external and progressive internal fouling model fitting to the experimental data obtained in runs 4, 6 and 8 (TMP = 1 bar; cross flow velocity = 3 m/s).

Fig. 7 shows the pore narrowing model fitting to the experimental data obtained in runs 4, 6 and 8. This model does not fit to the experimental data well (the values obtained for t and $1/Q_f^{0.5}$ from the experimental data do not form straight lines as the model predicts and implies) and implies that pore narrowing model is not suitable to predict the performance of the membrane in clarifying sugar cane juice.

5.3.3. Combination of external and progressive internal fouling

In this model, the cake filtration model is modified to include an increase in the specific cake resistance due to pore narrowing. Thus, the following expression could be used in this model:

$$Q_f = \frac{P_{tm}A_o}{\mu_f\{R_{m0} + (\alpha C_W/A_0 + 2C/V_p)V_f\}} \quad (12)$$

where P_{tm} is the transmembrane pressure and μ_f is the viscosity of the permeate. According to this model, a plot of $1/Q_f$ against V_f should yield a straight line as given in Eq. (12).

$$\frac{1}{Q_f} = \left[\frac{\mu_f}{P_{tm}\,A_o}\right]\left(\frac{\alpha C_W}{A_0} + \frac{2C}{V_p}\right)V_f + \left[\frac{\mu_f R_{m0}}{P_{tm}\,A_o}\right] \quad (13)$$

Fig. 8 shows the combination of external and progressive internal fouling model fitting to the experimental data obtained in runs 4, 6 and 8. While this model fits well with the data obtained in all three runs mentioned above, the initial flow rate or flux predicted by the model fits well when the pore size of the membrane becomes larger. This indicates that the combination of external and progressive internal fouling model is much suitable for microfiltration membranes but could be used to predict the performance of ultrafiltration membrane with reasonable accuracy as shown in Table 8.

5.3.4. Complete pore blocking model

This considers the drastic form of internal fouling when the particle sizes are comparable to the membrane pore size where they could completely block the pores. Thus the surface area of the membrane, A is reduced over the time as given below:

$$A = A_o - \sigma V_f \quad (14)$$

where σ is a parameter characterising the plugging potential of the suspension which is proportional to the concentration of particles in the feed solution. Thus the flow rate, Q_f at a given time, t can be given by:

$$Q_f = Q_o\ \exp(-\sigma J_o t) \quad (15)$$

where J_o is the initial permeate flux. Eq. (7) can be rearranged as below to get a linear plot for $\ln[Q_f]$ against t:

$$\ln\ [Q_f] = \ln[Q_o] - \sigma J_o t \quad (16)$$

Fig. 9 shows the complete pore blocking model fitting to the experimental data obtained in runs 4, 6 and 8. This model does not fit to the experimental data well (the values obtained for t and $\ln[Q_f]$ from the experimental data do not form straight lines as the model predicts) and implies that complete pore blocking model is not suitable to predict the performance of the membrane in clarifying sugar cane juice.

The combination of external and progressive internal fouling model indicates that 0.1 μm membrane fouls both on the surface (cake fouling) as well as the internal pores. Thus, fouled pores receive better cleaning during the air back flushing which improves the flux. However, the cake filtration model suggests that both the 0.02 and 0.05 μm membranes foul more on the surface compared to the internal pores. The air back flushing was not efficient in removing the surface fouling of those membranes. Smaller pore sizes could have reduced the efficiency of surface cleaning by the air. This needs further investigation. Further, in terms of modeling the fouling of membrane, similar results were obtained by Jacob and

Table 8
Combination of external and progressive internal fouling model fitting to the experimental data.

Run no.	Membrane pore size (μm)	Model input and output		Experimental value of $[\mu_f R_{m0}/P_{tm}A_o]$ (h/L)	Error in the value of $[\mu_f R_{m0}/P_{tm}A_o]$ (%)
		R_{m0} (m^{-1}) (input)	$[\mu_f R_{m0}/P_{tm}A_o]$ (h/L) (output)		
4	0.10	7.76×10^{11}	0.0412	0.0485	15.1
6	0.05	5.98×10^{11}	0.0318	0.0594	46.5
8	0.02	1.16×10^{12}	0.0617	0.0507	−21.7

Transmembrane pressure, $P_{tm} = 1 \times 10^5$ Pa; A_o = membrane surface area, 0.1193 m^2; membrane porosity, 0.35; viscosity of sugar juice, μ_f is assumed as 0.798×10^{-3} Pa s.

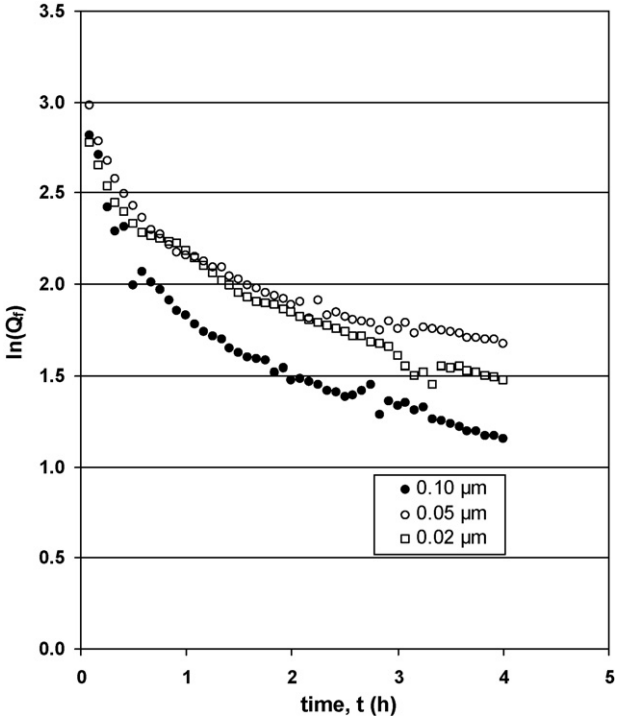

Fig. 9. Complete pore blocking model fitting to the experimental data obtained in runs 4, 6 and 8 (TMP = 1 bar; cross flow velocity = 3 m/s).

Jaffrin [13] for ultrafiltration. The cake filtration model fitted well for the ultrafiltration membrane (15 kDa) in their study which was comparable to our results. In the case of microfiltration (0.1 μm) the cake filtration model was unsatisfactory and the pore narrowing model provided better fit to their experimental results. However, in our study, combination of external and progressive internal fouling fits the microfiltration experiments well. This is possibly due to the difference in particle size distribution of the feed solutions used in both studies.

6. Conclusions

The performance of ceramic membranes with pore sizes of 0.02, 0.05 and 0.10 μm in clarifying sugar cane juice was investigated under different operating conditions and the following results were obtained from this study:

(1) For the 0.10 μm membrane, the increase in transmembrane pressure (TMP) from 1 to 3 bar increased the initial flux from 55.6 to 64.2 L/(m^2 h) (15.5% increase) and increased the average flux over a period of 4 h from 35.2 to 39.4 L/(m^2 h) (11.9% increase). The membrane was chemically cleaned before each of those experiments with 2% HNO_3 solution for 40 min followed by 2% NaOH for another 40 min. Although the intrinsic resistance of the membrane, R_{m0} returned closer to the initial R_{m0}, they were in the range of 3.87×10^{12} to 4.12×10^{12} m^{-1}, which seems to be 10-fold higher compared to the standard intrinsic resistance value of a 0.10 μm ceramic membrane.
(2) The initial flux of the 0.1 μm membrane increased dramatically to 140.7 L/(m^2 h) at a TMP of 1 bar when the membrane underwent a chemical cleaning with 1% NaOH with 3000 ppm free chlorine for 1 h and the average flux over a period of 4 h was also increased to 46 L/(m^2 h). The corresponding R_{m0} was 0.78×10^{12}.
(3) Among the three membranes tested 0.05 μm membrane performed better than the other two membranes and yielded initial and average fluxes of 165.4 and 65.6 L/(m^2 h) at a TMP of 1 bar. While the initial and average fluxes of 0.02 μm membrane were 135.0 and 36.6 L/(m^2 h) under the same TMP.
(4) Out of the four fouling models used to fit the experimental data, the cake filtration model fitted the performance of all membranes but predicted the initial fluxes of 0.02 and 0.05 μm membrane more accurately. This implies the cake filtration become dominant when the membranes have pore sizes in the ultrafiltration range where internal membrane pore blocking becomes insignificant.
(5) On the other hand, the combination of external and progressive internal fouling model also fitted the performance of all membranes but predicted the performance of 0.1 μm membrane better compared to the others. This implies that the membrane that has pore size in the microfiltration range is susceptible to both external as well as progressive internal fouling.
(6) Intermittent air back flushing improved the performance of 0.10 μm but did not have any effect on the performance of the other two membranes. This again emphasize the type of fouling that occurs on the membranes with different pore sizes and indicates that the intermittent air back flushing is effective in reducing internal pore blocking but not the fouling due to the cake layer that forms on the external surface of the membrane.
(7) However, all the membranes produced high quality filtered juice with more than 1.7 unit of purity rise, 99.6% reduction in turbidity and 38.9% reduction in color.

Thus, clarifying limed and partially treated sugar cane juice can be achieved successfully using micro- and ultrafiltration ceramic membranes. The membranes are capable of producing average fluxes in the range of 46–66 L/(m^2 h) for an operating period of 4 h when processing the above juice at 60 °C. Recovery of the membranes at the end of a filtration cycle could be carried out effectively through chemical cleaning; 1% NaOH and 3000 ppm of free chlorine solution is used for an hour to clean the membrane and to bring the intrinsic membrane resistance back closer to the original value. Fouling of the ultra- and microfiltration membranes could be expressed better by cake filtration model and combination of external and progressive internal fouling model, respectively. Further studies are warranted to select appropriate pore size of ceramic membrane to treat limed and partially clarified sugar cane juice.

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